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Japanese Patent Application JP 2002 - 53741 A

**Processability Improving Agent for PET-G Resins and  
PET-G Resin Composition**

Your Ref: 103003 - 07

For: Eastman Chemical Company

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**(54) [Title of the Invention]**

**Processability Improving Agent for PET-G  
Resins and PET-G Resin Composition**

**(57) [Summary]**

**[Object]** To provide an improving agent that produces excellent calenderability without reducing the transparency of the resulting PET-G product, and a PET-G resin composition containing this improving agent.

**[Means of Achievement]** A processability improving agent for PET-G resins, characterized in comprising as its principal component a single ester or a mixture of two or more esters composed of a polybasic acid and an aliphatic compound having hydroxyl groups in the molecules thereof, wherein at least one free carboxyl group remains per ester molecule; and a PET-G resin composition containing the processability improving agent.

**[Claims]**

**[Claim 1]** A processability improving agent for PET-G resins, characterized in comprising as its principal component a single ester or a mixture of two or more esters composed of a polybasic

acid and an aliphatic compound having hydroxyl groups in the molecules thereof, wherein at least one free carboxyl group remains per ester molecule.

**[Claim 2]** The processability improving agent for PET-G resins according to claim 1, characterized in that the aliphatic compound having hydroxyl groups in the molecules thereof is a partial ester of a higher fatty acid and higher aliphatic monohydric alcohol, polyhydric alcohol with 2 to 6 carbons, and/or the polycondensation product thereof.

**[Claim 3]** The calenderability improving agent for PET-G resins according to claim 1 or 2, characterized in that the polybasic acid is an aliphatic dicarboxylic acid or tricarboxylic acid.

**[Claim 4]** A PET-G resin composition, characterized in that it comprises the processability improving agent according to claim 1, 2, or 3.

#### **[Detailed Description of the Invention]**

##### **[0001]**

**[Technological Field of the Invention]** The present invention relates to a molding processability improving agent used, e.g. for the calendering of PET-G resins, and a PET-G resin composition containing this improving agent.

##### **[0002]**

**[Prior Art]** Polyester resins such as polyethylene terephthalate (PET) are widely used as container and packaging materials for food products and medical products; cases and molded container lids for electronic devices and office equipment; and ID card sheet.

**[0003]** Molding of these polyester resin films and sheets is generally performed by means of extrusion. Nevertheless, extrusion molding is slow, and does not yield a good level of productivity. On the other hand, calendering is a very productive method for molding film and sheet that has conventionally been used for vinyl chloride resin sheet and the like.

**[0004]** It is necessary to thoroughly melt the resin when molding film and sheet by means of calendering, but polyester resin will tightly adhere to the calender rolls once in a molten state, making calendering difficult. When melting is controlled and the calendering temperature is reduced, the resin will have better adhesion with regard to the surface of the rolls, but problems will be encountered with the rough surface of the molded film or sheet, resulting in an unsatisfactory product.

[0005] PET resin is a polyester resin that generally enjoys widespread use as a polyester resin, but PET resin has a high degree of crystallinity, and is therefore difficult to fashion into film or sheet by means of calendering.

[0006] Glycol-modified PET (referred to as "PET-G" in the present specification), which is a modified amorphous copolyester wherein part of the ethylene glycol component of the polyethylene terephthalate has been replaced by cyclohexane dimethanol, has been developed and marketed as a resin that alleviates the disadvantages of PET resins attributed to their crystallinity, although the problem persists that the polyester resin adheres to the roll surface during calendering, which is a characteristic of such resins.

[0007] The use of butyl stearate, cetyl palmitate, stearic acid monoglyceride, stearic acid diglyceride, stearic acid triglyceride, montan acid ester, montan acid ester partially saponified with calcium, wax ester, or aliphatic dicarboxylic acid ester, or other fatty acid ester lubricant with a copolyester wherein the ethylene glycol component of polyethylene terephthalate resin has been replaced with 10 to 40 mol% of cyclohexane dimethanol has been disclosed as a method for alleviating problems encountered when calendering using this type of polyester resin (JP (Kokai) 11-343353). Nevertheless, problems persist; namely, the fatty acid esters disclosed in the specification thereof yield insufficient results, adequate roll release properties cannot be obtained, and the sheet becomes cloudy when large amounts of the ester are added in order to obtain the release properties.

[0008] The need therefore exists for a processability improving agent for polyester resins that has good roll release properties and does not reduce transparency.

[0009]

**[Problems to Be Solved by the Invention]** It is an object of the present invention to provide an improving agent that produces excellent calenderability without reducing the transparency of the resulting PET-G product, and a PET-G resin composition containing this improving agent.

[0010]

**[Means Used to Resolve the Above-Mentioned Problems]** As a result of having conducted diligent studies in order to resolve the above-mentioned problems, the inventors perfected the present invention upon discovering that an ester comprising a polybasic acid and an aliphatic compound having hydroxyl groups in the molecules thereof, wherein at least one free carboxyl

group remains per ester molecule, produces excellent calenderability without reducing transparency of the PET-G product.

[0011] In other words, the present invention is constituted as follows.

1. A processability improving agent for PET-G resins, characterized in comprising as its principal component a single ester or a mixture of two or more esters composed of a polybasic acid and an aliphatic compound having hydroxyl groups in the molecules thereof, wherein at least one free carboxyl group remains per ester molecule.

[0012]

2. The processability improving agent for PET-G resins according to (1) above, characterized in that the aliphatic compound having hydroxyl groups in the molecules thereof is a partial ester of a higher fatty acid and higher aliphatic monohydric alcohol, polyhydric alcohol with 2 to 6 carbons, and/or the polycondensation product thereof.

[0013]

3. The processability improving agent for PET-G resins according to (1) or (2) above, characterized in that the polybasic acid is an aliphatic dicarboxylic acid or tricarboxylic acid.

[0014]

4. A PET-G resin composition, characterized in that it comprises the processability improving agent according to (1), (2), or (3) above.

[0015]

**[Embodiments of the Invention]** The present invention shall now be described in detail. The resin that is an object of the present invention is a modified amorphous copolyester (PET-G) in which part of the ethylene glycol component of the polyethylene terephthalate has been substituted by cyclohexane dimethanol.

[0016] Examples of "aliphatic compounds having hydroxyl groups in the molecules thereof" constituting the ester of the present invention, which is composed of a polybasic acid and aliphatic compound having hydroxyl groups in the molecules thereof, include aliphatic monohydric alcohols such as lauryl alcohol, myristyl alcohol, cetyl alcohol, stearyl alcohol, arachyl alcohol, and behenyl alcohol; alcohols with two or more hydroxyl groups such as ethylene glycol, 1,2-propanediol, 1,3-propanediol, 1,4-butanediol, 2,3-butanediol, 1,5-pentanediol, 1,6-hexanediol, glycerin, trimethylolpropane, erythritol, pentaerythritol,

dipentaerythritol, xylitol, mannitol, sorbitol, and sorbitan; and/or polycondensation products thereof. Alternative examples include or partial esters of these polyhydric alcohols and an aliphatic carboxylic acid such as caprylic acid, capric acid, lauric acid, myristic acid, palmitic acid, stearic acid, behenic acid, oleic acid, linolic acid, linolenic acid, and erucic acid.

[0017] Examples of the "polybasic acid" constituting the ester of the present invention, which is composed of a polybasic acid and an aliphatic compound having hydroxyl groups in the molecules thereof, include aliphatic dicarboxylic acids such as oxalic acid, malonic acid, succinic acid, glutaric acid, adipic acid, pimelic acid, suberic acid, azelaic acid, sebacic acid maleic acid, fumaric acid, citraconic acid, itaconic acid, malic acid, and tartaric acid; and aliphatic tricarboxylic acids such as tricarballylic acid and citric acid.

[0018] The ester of the present invention, which is composed of a polybasic acid and an aliphatic compound having hydroxyl groups in the molecules thereof, may be prepared by means of performing an esterification with a known method, using a single ester or a mixture of two or more esters of the above-mentioned polybasic acid and an aliphatic compound having hydroxyl groups in the molecules thereof, such as an alcohol. It is also possible to use these reactions products alone or as a mixture.

[0019] The processability improving agent for PET-G resins pertaining to the present invention comprises a single ester or a mixture of two or more esters, but may additionally contain other components as long as the single ester or mixture of esters is its principal component.

[0020] In addition, at least one carboxyl group derived from a polybasic acid in the reaction product must remain in the ester of the present invention, which comprises the polybasic acid and an aliphatic compound having hydroxyl groups in the molecules thereof. The effect of improving the polyester resin, which is an object of the present invention, will not be realized as a result of using an ester that has been obtained by means of using all of the carboxyl groups in the polybasic molecules as the ester in the reaction.

[0021] The admixing of a polyester resin with the ester in the present invention, which comprises a polybasic acid and an aliphatic compound having hydroxyl groups in the molecules thereof, improves the adhesion of PET-G resin compositions on the calender surface during calender molding, resulting in a product having excellent transparency.

[0022] An appropriate amount of the ester of the present invention, which comprises a polybasic acid and an aliphatic compound having hydroxyl groups in the molecules thereof, is within a

range of 0.1 to 3.0 parts by mass<sup>TN1</sup> per 100 parts by mass of PET-G. If the amount added is less than 0.1 parts by mass, the adhesion of the resin to the calender roll surface will not be sufficiently alleviated, while if the amount added is greater than 3.0 parts by mass, the product may occasionally suffer from reduced transparency.

[0023] There are no particular limitations as to the method for admixing the PET-G resin with the ester of the present invention, which comprises a polybasic acid and an aliphatic compound having hydroxyl groups in the molecules thereof, as long as these components may be uniformly mixed; for example, mixing may be performed with a variety of mixing machines, such as a Banbury mixer or heating rolls.

[0024]

**[Working Examples]** The present invention will now be described with working examples; but the present invention shall not be limited to these working examples.

[0025] (Samples for testing)

1. Glycerol succinate laurate

Succinylated glycerin laurate was obtained by means of a well-known esterification method using one mole of glycerin monolaurate (Riken Vitamin Co., Ltd.; distilled monoglyceride: Rikemal M-300) and one mole of succinic anhydride.

[0026] 2. Glycerol succinate stearate

Succinylated glycerin stearate was obtained by means of a well-known esterification method using one mole of glycerin monostearate (Riken Vitamin Co., Ltd.; distilled monoglyceride: Rikemal S-100) and one mole of succinic anhydride.

[0027] 3. Glycerol succinate behenate

Succinylated glycerin behenate was obtained by means of a well-known esterification method using one mole of glycerin monobehenate (Riken Vitamin Co., Ltd.; distilled monoglyceride: Rikemal B-100) and one mole of succinic anhydride.

[0028] 4. Glycerol citrate laurate

Citratized glycerin laurate was obtained by means of a well-known esterification method using one mole of the above-mentioned glycerin monolaurate and one mole of citric acid.

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<sup>TN1</sup> **Translator's Note:** The Japanese source text uses the Japanese characters for "mass" as opposed to "weight." Therefore, we have decided to reflect this choice made by the Japanese authors of this patent, although the more conventional English expression would probably be "parts by weight."

[0029] 5. Glycerol citrate stearate

Citratred glycerin stearate was obtained by means of a well-known esterification method using one mole of the above-mentioned glycerin monostearate and one mole of citric acid.

[0030] 6. Glycerol citrate behenate

Citratred glycerin behenate was obtained by means of a well-known esterification method using one mole of the above-mentioned glycerin monobehenate and one mole of citric acid.

[0031] 7. Stearyl citrate

Stearyl citrate was obtained by means of a well-known esterification method using one mole of citric acid and two moles of stearyl alcohol (Kao Corporation: Kalcol 8688).

[0032] 8. Polyethylene wax (Mitsui Petrochemical Industries, Ltd.; Hiwax 200P)

[0033] 9. Stearic acid (Nippon Oil & Fat Co., Ltd.; horse/beef tallow stearic acid)

[0034] (Tests and Evaluation)

A specific amount (parts by mass) of sample was admixed per 100 parts by mass of PET-G resin (Eastman Chemical Company; Easter PET-G6763), kneaded with a twin-screw extruder, and pelletized. The resulting pellets were heated to 155°C, kneaded for five minutes with an 8-inch roll, and molded into sheet with a thickness of 2 mm. The roll release properties exhibited during molding were observed and evaluated. The resulting sheets were pressed for five minutes at 180°C and 130 Mpa to make pressed sheets with a thickness of 5 mm. The transparency was then macroscopically evaluated. The evaluations were based on the following criteria.

[0035] (Release properties)

A: Released smoothly without adhering to the roll.

B: Adhered slightly to roll.

C: Adhered to roll, and suffered poor release.

[0036] (Transparency)

A: Good transparency

B: Fair transparency

C: Poor transparency

[0037]

[Table 1]

|                       | Sample                      | Amount added | Release properties | Transparency |
|-----------------------|-----------------------------|--------------|--------------------|--------------|
| Working Example 1     | Glycerol succinate laurate  | 2.0          | A                  | A            |
| Working Example 2     | Glycerol succinate stearate | 0.5          | A                  | A            |
| Working Example 3     | Glycerol succinate behenate | 0.3          | A                  | A            |
| Working Example 4     | Glycerol citrate laurate    | 1.0          | A                  | A            |
| Working Example 5     | Glycerol citrate stearate   | 1.0          | A                  | A            |
| Working Example 6     | Glycerol citrate behenate   | 0.1          | A                  | A            |
| Working Example 7     | Stearyl citrate             | 0.5          | A                  | A            |
| Comparative Example 1 | Glycerin monostearate       | 1.0          | C                  | -            |
| Comparative Example 2 | Glycerol succinate stearate | 0.05         | B                  | -            |
| Comparative Example 3 | Stearyl citrate             | 5.0          | A                  | B            |
| Comparative Example 4 | Polyethylene wax            | 0.3          | A                  | C            |
| Comparative Example 5 | Stearic acid                | 0.3          | B                  | A            |

[0038]

[Effect of the Invention] The admixing of the improving agent of the present invention will result in good calenderability, and a worked PET-G product with excellent transparency.